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Influence irradiation by high energy heavy ions on piezoelectric, elastic and dielectric properties of ZnO single crystals

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ABSTRACT : C^{12} (90 MeV) and Ne^{22} (160 MeV) ion irradiation effects on dielectric, piezoelectric and electromechanical properties of ZnO single crystals have been studied. It was shown that high energy heavy ion irradiation leads to significant changes in electromechanical properties of piezoelectric materials.

1 - INTRODUCTION

The last few years, high energy ion accelerators became available for solid state study and now high energy ion implantation is one of the most perspective methods of structural modification of different solids. At variance from low energy ion irradiation, the dominating electron stopping mechanisms of the energetic ions is the physical basis of the high energy ion implantation effects the most important of which seem to be selfannealing of the radiation damage, the increase of the defect thermal stability, the impurity migration along the ion tracks and the formation of high pressure local regions [1, 2]. For energy more than 1 MeV/a.m.u. ion ranges can exceed dozens and hundreds of micrometers which permit to change the properties of bulk irradiated materials. In ref. [3], the nature of radiation damages in ZnO single crystals after 90 MeV carbon and 110 MeV oxygen ions bombardment was investigated by using optical spectroscopy. The present report deals with the first results of experimental investigations of electromechanical properties of ZnO irradiated with 90 MeV C^{12} and 160 MeV Ne^{22} ions.

2 - EXPERIMENTAL AND RESULTS

Crystals were grown by hydrothermal method in alkaline media containing LiOH [4]. The samples were prepared as X-cut (001O) and Z-cut (0001) plates ($5 \times 6 \times 0.6 \text{ mm}^3$) and Z-cut bars ($9 \times 2 \times 1 \text{ mm}^3$). In order to increase the electrical resistivity, all samples were annealed in lithium carbonate melting [4].

The irradiation was performed with extracted on air carbon and neon ion beams on U-200 and U-400 accelerators of cyclotron complex of Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna. The first series of samples was irradiated with carbon ions and after measurements was additionally irradiated with neon ions. The temperature of crystals during irradiation never exceeded 100°C .

In order to identify the resonances giving either thickness longitudinal vibrations of Z-cut plates, or thickness shear vibrations of X-cut plates or XZOl bar vibrations, spectra of elastic vibrations were measured before and after every stage of the ion irradiation.

Elastic moduli C_{33}^D , C_{44}^D and elastic constants S_{11}^E were calculated from expressions :

$$f_{aR} = (f_{44}^D \rho)^{1/2}, \quad f_{aR} = (f_{33}^D \rho)^{1/2}, \quad f_R = (S_{11}^E \rho)^{-1/2},$$

where f_R and f_{aR} - resonance and antiresonance frequencies, t -plates thickness, ρ -density. Dielectric constants at invariable mechanical stress and invariable deformation ϵ_{11}^T , ϵ_{33}^T , ϵ_{11}^S and ϵ_{33}^S were measured for frequencies 1 KHz and 10 MHz. The knowledge of resonance and antiresonance frequencies allows to determine electromechanical coupling factors of the investigated vibrations and, further, from

these factors, elastic modula, dielectric constants are used to obtain piezoelectrical constants e_{33} , e_{15} and piezoelectrical modulus d_{31} by using expressions :

$$e_{33} = K_t \cdot (C^{D_{33}} \cdot \epsilon D_{33})^{1/2}, \quad e_{15} = K_{15} \cdot (C^{D_{44}} \cdot \epsilon S_{11})^{1/2}$$

$$d_{31} = K_{31} \cdot (S^{E_{11}} \cdot \epsilon T_{11})^{1/2}$$

Table 1 : The bulk resistance $\rho(\Omega.cm)$, dielectrical constants ϵT_{11} , ϵS_{11} , ϵT_{33} , ϵS_{33} , resonance frequencies f_R (KHz) and Q-factors of ZnO crystals before and after ion irradiation. The relative accuracy of frequency measurements- .0010.

Parameter	Before irrad.	C12 ion irrad.	C12 and Ne22 ion irrad.	Lit. data [5, 6]
ρ_1	$3.79.10^{10}$	$0.54.10^{10}$	$1.4.10^{11}$	
ρ_2	$(1.9-18.1).10^{11}$	$(9.0-0.57).10^{10}$	$(10-6).10^{11}$	
$\epsilon T_{11}/\epsilon_0$	9.13	9.3	8.48	9.26 8.67
$\epsilon S_{11}/\epsilon_0$	8.68	8.83	8.1	8.33 8.33
$\epsilon T_{33}/\epsilon_0$	11.26	11.28	10.66	11 11.26
$\epsilon S_{33}/\epsilon_0$	9.26	10.7	9.97-9.26	8.84 8.81
f_R sl. Z cut	3578.002	3522.001	3618.972	
f_R sl. X cut	1735.889	1723.502	1742.355	
f_R	260.885	260.765	262.033	
Q	2358	4575	8188	

Furthermore, an analyze of elastic vibration spectra permits also to estimate or to measure by usual methods the Q-factor of corresponding resonators. Besides mentioned above parameters, the electroconductivity of samples was also measured. It should be noted that measurements were done at conditions when all samples were isolated from light at least twenty-four hours.

Experimental data are given in tables 1, 2 and consist for comparison the litterature data about unirradiated crystals were obtained by hydrothermal method [5, 6].

Table 2 : Elastic modula $C^{D_{33}}$, $C^{D_{55}}$ ($N.m^{-2}$), elastic compliancies $S^{E_{11}}$ ($m^2.N^{-1}$, piezoelectric modulus d_{31} (CGSE), piezoelectric constants e_{33} and e_{15} , electromechanical coupling factors K_t , K_{15} , K_{31} before and after irradiation.

Parameter	Before irrad.	C12 ion irrad.	C12 and Ne22 ion irrad.	Lit. data [5, 6]
$C^{D_{33}}$	$23.79.10^{10}$	$23.11.10^{10}$	$23.34.10^{10}$	$21.50.10^{10}$ $22.10.10^{10}$
$C^{D_{55}}$	$4.74.10^{10}$	$4.66.10^{10}$	$4.77.10^{10}$	$4.72.10^{10}$ $4.48.10^{10}$
$S^{E_{11}}$	$78.89.10^{-13}$	$78.96.10^{-13}$	$78.28.10^{-13}$	$79.10.10^{-13}$
d_{31}	$15.57.10^{-8}$	$15.47.10^{-8}$	$15.43.10^{-14}$	$15.43.10^{-8}$ $15.36.10^{-8}$
e_{33}	$33.6.10^4$	$28.0.10^4$	$34.00.10^4$	$34.20.10^4$ $28.8.10^4$
e_{15}	$12.97.10^4$	$12.85.10^4$	$11.67.10^4$	$17.7.10^4$ $11.1.10^4$
K_t	$25.37.10^{-2}$	$19.44.10^{-2}$	$25.34.10^{-2}$	$28.2.10^{-2}$ $22.9.10^{-2}$
K_{15}	$22.68.10^{-2}$	$22.45.10^{-2}$	$21.04.10^{-2}$	$31.5.10^{-2}$ $19.9.10^{-2}$
K_{31}	$18.46.10^{-2}$	$18.33.10^{-2}$	$18.36.10^{-2}$	$18.9.10^{-2}$ $18.10.10^{-2}$

3 - DISCUSSION

3.1 - Dielectric properties - As seen from table 1, dielectric constants of ZnO irradiated by C^{12} have been increased (the most essential variation is for ϵ_{S33} , measured at frequency 10 MHz). The next irradiation by Ne^{22} ions has led to the decrease of dielectric constants, which is manifested at low frequencies (1 KHz) much stronger and quite weakly changed at the 10 MHz for the value ϵ_{S33} .

The bulk resistance of crystals irradiated by C^{12} ions in both directions (along 6 axis and perpendicular to it) decreases (ρ_1 and ρ_2), but it is practically restored by a second irradiation by Ne^{22} ions.

3.2 - Elastic properties - Resonance frequencies are substantially influenced by the irradiation (they decrease at irradiation by C^{12} ions and increase above original ones by a second Ne^{22} ions irradiation). If we assume the density of ZnO to be constant, then such essential variations can be only due to the decrease of crystal stiffness after carbon irradiation and increase of stiffness after neon irradiation. Similar result has been obtained for quartz irradiation by neutrons (increasing of resonance frequency of the AT Z-cut resonators. So, at dose of $1.2 \cdot 10^{18}$ neutron.cm $^{-2}$ the relative increase of the resonator frequency was .10 [7]. In our case, for ZnO bar, one has detected the decrease of frequency to .0460 at the first irradiation and the increase of frequency to .4380 by second irradiation.

Data of f_R = variation are given in table 3.

Table 3 : Resonance frequencies of ZnO irradiated with high energy heavy ions and SiO $_2$ after neutron irradiation [7].

Crystal	Orientation	Type of irradiation	Frequency change	
SiO $_2$	At-cut	neutron $1.2 \cdot 10^{18}$ cm $^{-2}$	increase	0.10
ZnO	bar	ions C^{12}	decrease	0.0460
	X-cut	ions C^{12}	decrease	0.7130
	Z-cut	ions C^{12}	decrease	1.560
	bar	ions C^{12} + Ne^{22}	increase	0.4380
	X-cut	ions C^{12} + Ne^{22}	increase	0.370
	Z-cut	ions C^{12} + Ne^{22}	increase	1.140

3.3 - Q-factor - Q-factor is the measurement of the internal friction as the rest channels of the elastic energy loosing prior and after irradiation have not been changed. In fig. 2 you can see a part of amplitude frequency characteristics of the element performing longitudinal vibration near the main resonance. This indicates the manner in which the resonance frequency decreased after carbon ion irradiation, but the Q-factor increased. After irradiation by neon ions the resonance frequency essentially increased and the Q-factor increased more significantly (see table 1).

3.4 - Factor of electromechanical coupling - Factor K_t responsible for excitation of thickness longitudinal vibrations of Z-cut plates essentially decreases after irradiation by C^{12} ions. The rest factors vary but slightly. After the second irradiation K_t is restored and approaches its original value (Table 2).

3.5 - Piezoelectric moduli and constants - Piezoelectric modulus d_{31} is practically not changed, but piezoelectric constant e_{15} decreases both at C^{12} ion irradiation and next irradiation by Ne^{22} ions. At the same time, piezoelectric constant e_{33} more significantly decreases at irradiation by C^{12} ions and is restored to its original value after irradiation by Ne^{22} ions. From the data obtained, one can conclude that variation of electromechanical coupling factors is mainly determined by variation of piezoelectric constants rather than by dielectric constants and elastic moduli. One should note, that crystals used in our work display their properties : values of piezoelectric constants and electromechanical coupling factors are intermediate between those of crystals described in the works [5, 6]. It seems that indicated differences are due to different impurity contamination.

To summarize, the data given in tables 1-3 and their next discussion show that irradiation of ZnO by C^{12} ions leads to the increase of quality of resonators independently on irradiation by neon ions. A high electrical resistivity and piezoproperties are kept constant and quality of resonators making longitudinal vibrations along X essentially increase when excited electrical field is applied along Z. However, in the last case the spectrum of elastic vibrations is complicated due to additional resonances, but on the basis of the data obtained, one cannot connect it to the total increase of the quality.

Probably, radiation effects observed in ZnO crystals are associated with structural modifications that take place at irradiation and also with the processes of redistribution of impurities. A possible process of redistribution of impurities can be due to the regions with a mechanical stress and radiation stimulated diffusion which occurs in crystal during irradiation. Besides, it is very difficult to explain such significant changes of electromechanical properties as modifications in zone of ion range (some dozens of micrometers) at sample thickness of 1 mm order. One can assume that structural modifications and impurities redistribution happen to be beyond zone of ion range, that has been observed in LiF crystals [8]. To thoroughly define the processes proceeding in piezoelectric crystals at their irradiation by high energy heavy ions, one needs to carry out further investigations required for checking the above mentioned assumptions.

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